# Spontaneous anti-Stokes scattering as a high-resolution and picosecond-time-scale vuv light source[a)]

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A vuv and soft x-ray light source based on spontaneous anti-Stokes scattering from atomic population stored in a metastable level is proposed. It is shown that the source has a maximum brightness equal to that of a blackbody at a Boltzman temperature characteristic of the population of the metastable level. This maximum brightness is attained as the media approaches two-photon opacity. The source should have high resolution, may be of picosecond time scale, and on pulsed basis should be brighter than other laboratory-scale vuv light sources.

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There has recently been considerable interest in the possibility of constructing vuv and soft x-ray lasers by the technique of first inverting the population of a metastable species with respect to ground, and then extracting the inverted population by applying an intense laser tuned to the upper level of the resonance transition.[1,2] The operation of vuv laser sources of this type await the prior development of methods of accomplishing the inversion of the metastable species.

This letter proposes and describes a related type of vuv or soft x-ray light source which does not require an inversion of the metastable species. This source, though incoherent, is tunable, of very narrow linewidth, and may be of picosecond time scale. On a pulsed basis, its brightness and peak power (though not its ease and range of spectral coverage) should exceed that of other laboratory-scale light sources.

A schematic and energy-level diagram of the proposed light source is shown in Fig. 1. Population is stored in the metastable level of a selected species (for example, the $2s\,^1S$ level of He), with energy with respect to ground $\hbar\omega_2$. A tunable laser of frequency $\omega_p$ is focused to a confocal parameter equal to the desired source length $L$. Spontaneously emitted vuv radiation at frequencies $\omega_{uv}=\omega_2+\omega_p$ and $\omega_2-\omega_p$ is observed at right angles to the laser beam. The emitted radiation has a pulse length the same as that of the laser at $\omega_p$ and a linewidth which is the wider of the Doppler width of the $|1\rangle - |2\rangle$ transition, or of the incident laser.

To simplify and give a better physical interpretation to the formula, we assume a three-level nondegenerate atomic system (Fig. 1) with nonzero matrix elements $\mu_{23}$ and $\mu_{31}$.

In the presence of the laser pump field of frequency $\omega_p$ and electric field $E_p$, the per atom (per bandwidth) spontaneous emission rate at frequency $\omega$ may be written[3]

$$A(\omega)=\frac{\omega^3|\mu_{13}|^2}{3\pi\hbar\epsilon_0 c^3}\left[\sin^2\left(\frac{\mu_{23}E_p}{2\hbar\Delta\omega}\right)\right]g(\omega-\omega_{uv})$$

$$=\left(\frac{\omega}{\omega_{31}}\right)^3 A_{31}\left[\sin^2\left(\frac{\mu_{23}E_p}{2\hbar\Delta\omega}\right)\right]g(\omega-\omega_{uv}). \qquad (1)$$

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The quantity $A_{31}$ is the Einstein $A$ coefficient for spontaneous emission from level $|3\rangle$ to level $|1\rangle$. The line shape $g(\omega-\omega_{uv})$ denotes the narrow distribution of emitted radiation centered at the frequencies $\omega_{uv}=\omega_2\pm\omega_p$. The line shape is the convolution of the Doppler- or pressure-broadened linewidth of the $|1\rangle - |2\rangle$ transition, and the linewidth of the pumping laser, and is normalized so that $\int g(\omega-\omega_{uv})\,d\omega=1$. The quantity $\Delta\omega$ is defined as $\omega_{31}-\omega_{uv}$.

We see from Eq. (1) that if the laser pump field is adjusted so that $(\mu_{23}E_p/\hbar\Delta\omega)\rightarrow\pi$, the total emission rate per atom $\int A(\omega)\,d\omega\rightarrow(\omega/\omega_{31})^3A_{31}$. However, as we will see shortly, it will seldom be desirable to operate the light source at such high laser pump fields.

The key to optimizing the brightness of this light source is the two-photon absorption which is created at the ultraviolet frequency $\omega_{uv}$, in the presence of the laser pump frequency $\omega_p$. The cross section for two-photon absorption at $\omega$ may be written[3]

$$\sigma(\omega)=\frac{\pi\omega|\mu_{13}|^2}{3c\epsilon_0\hbar}\left[\sin^2\left(\frac{\mu_{23}E_p}{2\hbar\Delta\omega}\right)\right]g(\omega-\omega_{uv}) \qquad (2a)$$

$$=\frac{\pi^2c^2}{\omega^2}A(\omega). \qquad (2b)$$

We define the brightness of the light source $B(\omega)$ as the radiated power per area per steradian per band-

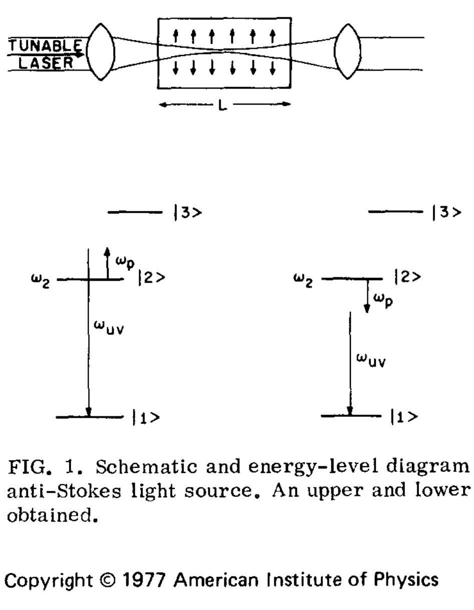


FIG. 1. Schematic and energy-level diagram for spontaneous anti-Stokes light source. An upper and lower sideband is obtained.

width. As the depth or thickness of the source is increased, by either increasing the gas pressure, the laser pump power density, or the radius of the laser beam, the brightness $B(\omega)$ tends to be increased as a result of the increased emission, but decreased by the corresponding increase in two-photon absorption. For an infinitely long cylinder, the variation of brightness with radius $r$ is described by

$$\frac{dB(\omega)}{dr} + \sigma(\omega)(N_1 - N_2)B(\omega) = \frac{\hbar\omega A(\omega)N_2}{4\pi} \quad (3)$$

with solution

$$B(\omega) = \frac{\hbar\omega A(\omega)}{4\pi\sigma(\omega)}\left(\frac{N_2}{N_1 - N_2}\right)\{1 - \exp[-\sigma(\omega)(N_1 - N_2)r_0]\}, \quad (4)$$

where $r_0$ is the outer radius of the focused (assumed cylindrical) laser beam. The total radiated power in a length $L$, per radian per sec of bandwidth is then $B(\omega)(2\pi)2\pi r_0 L$.[4]

We thus obtain the interesting result that the maximum source brightness will be approached when the incident laser pump power is increased to render the media nominally two-photon opaque, i.e., $\sigma(\omega)(N_1 - N_2) r_0 = 1$. The power density necessary to accomplish this is often much less than the $[(\mu_{23}E_p/\hbar\Delta\omega) = \pi]$ power density. Using Eq. (2b), the maximum brightness is then

$$B(\omega)_{\max} = \frac{1}{4\pi^3}\frac{\hbar\omega^3}{c^2}\frac{N_2}{N_1 - N_2} \quad (5a)$$

$$= \frac{1}{4\pi^3}\frac{\hbar\omega^3}{c^2}\left(\frac{1}{\exp(\hbar\omega_{21}/kT) - 1}\right). \quad (5b)$$

In Eq. (5b) we have used a Boltzman temperature $T$ such that $N_2/N_1 = \exp[-(\hbar\omega_{21}/kT)]$. We recognize Eq. (5b) as simply the brightness of a blackbody at temperature $T$.

When the source is optically thin, for example in a cw experiment where $\sigma(\omega)(N_1 - N_2)r_0 \ll 1$, Eq. (4) becomes $B(\omega) = [\hbar\omega A(\omega)N_2 r_0]/4\pi$. Since at a fixed laser power, $A(\omega) \sim 1/r_0^2$, the brightness is maximized by choosing $r_0$ as small as possible, and the total power emitted per bandwidth is invariant to $r_0$.

We now give a numerical example for storage in the $2s\,^1S$ level of $He$ at 166 278 cm$^{-1}$. The pertinent parameters[5] are the $A$ coefficient for the $2p\,^1P$-$1s^2\,^1S$ resonance transition, $A_{31} = 1.8 \times 10^9$ sec$^{-1}$; the matrix element $\mu_{23} = 2.9$ a.u.; and the Doppler width at 300 °K of the $1s$-$2s$ transition of 1.0 cm$^{-1}$.

The discharge conditions are less certain. As a benchmark for the operation of this device, we consider cw discharge conditions comparable to those of a He-Ne laser. For a 4-mm bore, 100 mA current, and 1 Torr He pressure, Silfvast has measured a $2s\,^1S$ population of $9 \times 10^{11}$ atoms/cm$^3$, yielding (at 300 °K) a ratio $N_2/N_1 = 2.8 \times 10^{-5}$ ($T = 22\,860$ °K).[6]

To minimize the requirements on laser pump power, it is desirable to tune $\omega_p$ near the $2s$-$2p$ transition frequency, but not so near that $\omega_{uv}$ is absorbed by ground-state atoms.[7] We assume a detuning of $\Delta\omega = 300$ cm$^{-1}$ $\cong 1$ Å. Thus $\omega_{uv} = 585.4$ Å and $\omega_p = 2.19\ \mu$. From Eq. (5), using $N_2/N_1 = 2.8 \times 10^{-5}$, we calculate a maximum brightness of $4.9 \times 10^{16}$ photons/sec/cm$^2$/sr/cm$^{-1}$ of bandwidth. This emission is only present during the time the pump laser is on and is attained only over the spectral region where the media is nominally (two-photon) opaque. From Eq. (2), the two-photon absorption cross section is $\sigma(\omega_{uv}) = 7.3 \times 10^{-24} P/A$ cm$^2$, with $P/A$ in W/cm$^2$. For a ground-state population $N_1 = 3.2 \times 10^{16}$ atoms/cm$^3$ and a laser beam waist of 100 $\mu$, the condition $\sigma N_1 r_0 = 1$ is attained at a laser power and power density of $6.8 \times 10^4$ W and $4.3 \times 10^8$ W/cm$^2$, respectively. For a 2-cm source length, the total emitted vuv power is 0.13 W, and the efficiency of conversion from laser to vuv power is $1.9 \times 10^{-6}$.

Though the maximum power per bandwidth is determined by the ratio $N_2/N_1$, the necessary laser pump power may be reduced by simultaneously increasing $N_1$ and $N_2$. The spectral range of two-photon opacity, and thus the total emitted power, may be increased by deliberately using a broadband pump laser or (if pump power is available) by increasing $r_0$.

Using high-peak-power ($\sim 10^6$ W) pump sources, a total tuning range (both upper and lower sideband) of perhaps 60 000 cm$^{-1}$ should be observable. The output power will vary sharply as each of the upper $np$ states are approached. Since vuv power is linear with incident laser power, the source may also be run cw, perhaps pumped with a cw He-Xe or He-Ar gas laser, and would have a linewidth of about 1/200 of the 584-Å He resonance line.[7]

Even for the conservative example chosen above, the brightness of this anti-Stokes source exceeds that of the He resonance line by about $10^3$. This inherent increase in brightness results from the population storage associated with the metastable level. Extension to shorter wavelengths, for example, storage in the $Li^+$ $2s$ level at 202 Å, can be accomplished by using spark radiation[8] to selectively photoionize inner shell electrons. Finally, we note that if the technical problem of the separation of source and sample can be solved, the source should have a sufficiently fast rise time and brightness to pump an inner shell ionized laser of the type proposed by Duguay and Rentzepis.[9,10]

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[10]A 584-Å source is of an appropriate wavelength to ionize the inner shell electron of Rb and create gain on the $5p^55s$-$4p^6$ 697-Å transition of $Rb^+$. A gain of $e^{10}$ requires a 1-m path length and an inversion of $4.6\times10^{11}$ atoms/cm$^3$.

# Instability of a low-pressure Na–noble-gas discharge

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A new type of discharge instability is reported for a low-pressure Na–noble-gas discharge ($\approx 5\times10^{-3}$ Torr Na; 5.5 Torr Ne-Ar, 1% Ar). The voltage-current ($V$-$I$) characteristic is shown to be multivalued in the voltage. The resulting instability is a discontinuity in the positive column which propagates in the direction of electron particle flow with velocity between about $10^5$ and $5\times10^5$ cm/s.

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Discharge instabilities[1] in weakly ionized atomic or molecular gases usually manifest themselves in the form of striations or constriction of the discharge. In this letter we report on a different type of instability, which occurs in a low-pressure Na—noble-gas discharge. The system used is a Na-Ne-Ar mixture with $\approx 5\times10^{-3}$ Torr Na, 5.5 Torr Ne-Ar (1% Ar) at 20 °C; a tube radius of 1 cm; and a tube length of 60 cm. The wall temperature ($T_w \approx 255$ °C) is controlled by means of a heat pipe oven.[2] The temperature homogeneity over the tube length was measured to be better than ± 0.2 °C. The instability consists of a discontinuity moving in the direction cathode to anode, separating a region where Ne radiation is observed, indicative[3] of a high electric field ($E$) and electron temperature ($T_e$), and a region where only Na radiation is observed, indicating a low $E$ and $T_e$. With increasing current ($I$) the former region expands at the cost of the latter. So, at one particular value of $I$ the discharge has two distinct $E$ values. This multivalued $E$-$I$ behavior was predicted earlier[2,4] but not yet confirmed experimentally. The propagation of the instability is also considered to be a novel feature in discharges of this type.

In this letter we report two experiments. First, time-dependent voltage-current, $V(t)$-$I(t)$, characteristics, measured with the circuitry of Fig. 1. The circuitry consists of a parallel resistance $R$ and a current source giving an externally controlled triangular current form $I_0(t)$, with rise time $\tau_r$ and amplitude $\Delta I_0$, superimposed on a bias current $I_b$. This combination becomes a voltage-stabilized source if $R$ tends to zero. The $V(t)$-$I(t)$ curves were displayed on an oscilloscope and photographed. An example is shown in Fig. 2(a). In the current interval between points K and L it is found that $\dot V(t) > 0$ while $\dot I(t) < 0$ (the dot indicates differentiation with respect to time). This implies that the $V(t)$-$I(t)$ curve is multivalued in $V$, called[5,6] voltage controlled [Fig. 2(b)]. Figure 3 shows results of $V(t)$-$I(t)$ curves for various values of $\dot I_0(t)$. Figure 3 shows that for the present situation the characteristics are multivalued for 4000 A/s $> \dot I_0(t) >$ 60 A/s for $R = 200\ \Omega$. It is noted, in contrast, that the $V$-$I$ curves from dc experiments [$\dot I_0(t) \to 0$] have been found to be single valued (see, e.g., Fig. 1.2 of Ref. 2).

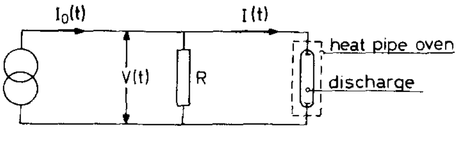


FIG. 1. Experimental setup (schematic).

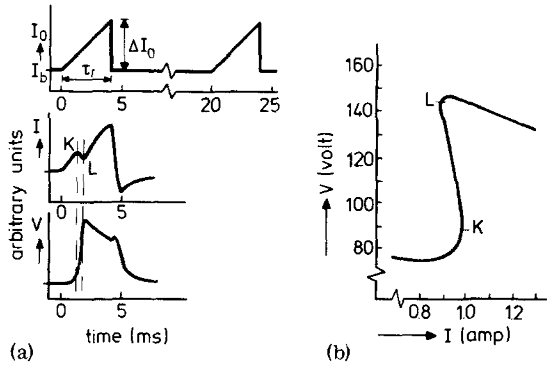


FIG. 2. Time behavior of total current $I_0(t)$, and oscilloscope traces of the resulting discharge current $I(t)$ and tube voltage $V(t)$. Conditions are $R = 200\ \Omega$, $\tau_r = 4$ ms, wall temperature $T_w = 254$°C, sodium wall density $n_w = 5\times10^{19}$ m$^{-3}$, $I_b = 0.7$ A, $\Delta I_0 = 1$ A, and $\dot I_0(t) = 250$ A/s. (b) Final result of the $V(t)$-$I(t)$ curve.